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New Data Evaluation Technique for Electron Tunneling Spectroscopy

by

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Submitted To

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A systematic study of Local Density Of States (LDOS) deconvolution from tip-surface tunneling spectra is reported. The one-dimensional WKB approximation is used to simulate the process. A new technique for DOS deconvolution from the electron tunneling spectroscopy data is proposed. The differential conductivity normalized to its fit to the tunneling probability function is used as a method of recovering sample DOS. This explicit procedure does not use unconstrained parameters and reveals a better DOS deconvolution in comparison with other techniques. The advantage of this new method is its feasibility for extracting two important physical parameters from experimental tunneling spectra: (i) local surface potential, and (ii) tip-sample distance. These values are the parameters used in the proposed fitting procedure. The local surface potential and the tip-sample distance retrieval are demonstrated by means of numerical simulations. Comparative Scanning Tunneling Spectroscopy (CSTS) is proposed as a new approach to eliminate the influence of the tip condition on the surface LDOS recovery.

I. Introduction

The first observation of the spectral density of electronic states in metal-oxide-superconductor tunneling experiments was made by Giaever.¹ This work led to a reexamination of the one-electron theory of tunneling that has dominated the field for more than 30 years². The new many-body transfer-Hamiltonian approach in tunneling current calculation was pioneered by Bardeen³ and developed by Cohen *et al.*⁴, Duke *et al.*⁵, Appelbaum and Brinkman⁶, Caroli *et al.*⁷ and many others.⁸ This formalism is used today in most applications of the tunneling theory.

The invention of Scanning Tunneling Microscopy (STM) by Binnig, Rohrer, Gerber and Weibel⁹ stimulated an additional interest in the theory of tunneling. Using the transfer-Hamiltonian formalism³, three-dimensional tunneling in the STM was studied by Tersoff and Hamann¹⁰, Garcia *et al.*¹¹, Feuchtwang *et al.*¹² and Lang.¹³

With the advent of STM, a Scanning Tunneling Spectroscopy (STS) with atomic spatial resolution became possible. The theory of STS was considered by Feuchtwang *et al.*¹², Selloni *et al.*¹⁴ and Lang.¹⁵ It was concluded that in the general case of three-dimensional tunneling, current *cannot be calculated* as a simple convolution of sample Densities Of States (DOS) and tip DOS with the "effective matrix element for tunneling". The simple relation between sample

Stroscio, Feenstra and Fein¹⁶ proposed an effective solution for the later problem. It was found that by normalization of the differential conductivity by the total conductivity one can effectively remove the dependence of the tunneling current on the tip-sample distance. The result was *qualitatively* generalized and it was concluded that this normalization procedure will cancel out both tip-sample distance and tip-sample voltage dependencies of the tunneling transmission probability in the differential conductivity.¹⁷ It was claimed that the $(dI/dV)/(I/V)$ function is “a relatively direct measure of the surface DOS”.¹⁶ This conclusion was confirmed by Lang¹⁵ in three-dimensional transfer-Hamiltonian numerical calculations for tunneling between Na and Ca single atoms positioned on infinite planar metal electrodes. The normalization technique was also justified by Hamers.¹⁸ Simmon’s formulas¹⁹ (WKB approximation) and constant DOS for the tip were used to simulate the tunneling current dependence on the tip-sample voltage. The recovered $(dI/dV)/(I/V)$ function has shown qualitative agreement with the input sample DOS. It is worthwhile to note that in this case¹⁸ less correspondence can be found between input and output sample densities of states in comparison with the “exact” simulation done by Lang.¹⁵ It is also important that in both calculations^{15,18} metal-like sample DOS were used.

Stroscio *et al.*¹⁶ experimentally tested the influence of the tungsten tip DOS on the deconvoluted sample density of states. The DOS of a Ni sample, deconvoluted by the $(dI/dV)/(I/V)$ technique, shows no tip-related features. This

conductivity (for details see Section III.1). Finally, Feenstra was able to obtain reproducible and reliable electronic spectra of III-V semiconductors.²³ However, with all these empirical adjustments, it is increasingly difficult to keep explicit the fundamental meaning of the normalization procedure as well as to justify its ultimate accuracy.

In this paper a new technique for DOS deconvolution from electron tunneling spectroscopy data is introduced. To recover the sample and/or tip DOS one may use the *differential conductivity normalized by its fit to the tunneling probability function*. The one-dimensional and semi-classical WKB approximation was used to model electron tunneling between the STM tip and the sample. As shown by simulations, in all studied cases this technique reveals better deconvolution of the input DOS in comparison with any existing technique.²³ This procedure does not use unconstrained parameters except the analytical form of the tunneling probability function. Since one should employ the same tunneling model to justify any other DOS deconvolution technique, the suggested procedure is more constrained and certainly more explicit than others.

The advantage of this new method is the feasibility of extracting two important physical parameters from experimental tunneling spectra: (i) "local work function," and (ii) tip-sample distance. These parameters are used in the proposed fitting of the differential conductivity by the tunneling probability function. The

The theory of electron tunneling spectroscopy has been discussed in many publications^{3,4,5,6,7,8,12,14,15}. Therefore, the goal of this section is to provide a short summary of the theory.

II.1 Methods of Tunneling Current Calculation.

The tunneling current between two weakly bounded electrodes using first-order perturbation theory is (refs. 6, 10, 12):

$$I(S, V) = \frac{2\pi e}{\hbar} \sum_{t,s} |M_{t,s}|^2 \delta(E_t - E_s) [\bar{f}(E_t - eV) - \bar{f}(E_s)], \quad (1)$$

where V is the sample bias with respect to the tip; S is the tip-surface separation, $M_{t,s}$ is the tunneling matrix element between states ψ_t of the tip and ψ_s of the surface; $\bar{f}(E)$ is the Fermi-Dirac distribution function; E_t and E_s are the energies of states ψ_t and ψ_s , respectively, in the absence of tunneling. Both energies are referenced to the surface Fermi level. Positive current indicates electron tunneling from tip to surface.

The tunneling matrix element can be calculated as follows using the method of Bardeen³:

$$M_{t,s} = -\frac{\hbar^2}{2m} \int (\psi_t^* \nabla \psi_s - \psi_s \nabla \psi_t^*) \cdot d\bar{A}, \quad (2)$$

where the integral is calculated over an arbitrary surface lying entirely within the vacuum region separating the tip and the surface.

The tunneling probability is a strong function of the parallel component of the energy $E_{||}$. At each particular total energy E , the DOS with zero $E_{||}$ component is heavily weighted by the tunneling probability in equation (3). Therefore, $\rho_s(E)$ and $\rho_t(E)$ may be approximated by the surface and the tip densities of electronic states with $k_{||} \approx 0$ (Ref. 18). This approximation may in part be a justification for ignoring the tunneling electron momentum in equation (3).

One can simplify equation (3) at low surface temperature, $kT \ll eV$ (k is Boltzmann's constant), by using the step function instead of the Fermi-Dirac distribution function. The density of the tunneling current is then:

$$J(S, V) \cong \frac{2\pi e}{\hbar} \left(\frac{\hbar^2}{2m} \right)^2 \int_0^{eV} T(S, V, E) \rho_s(E) \rho_t(E - eV) dE. \quad (5)$$

Assuming $E_{||} \approx 0$ for the electronic states which influence the tunneling current, the transmission probability can be written in the following form:

$$T(S, V, E) \cong \exp \left[-2S \sqrt{\frac{2m}{\hbar^2} \left(\bar{\Phi} + \frac{eV}{2} - E \right)} \right]. \quad (6)$$

Expressions similar to (5, 6) were employed in most cases in an attempt to describe semi-quantitatively the tunneling spectroscopy with spatial resolution.^{12,15,17,18,21} These equations were obtained for the weakly bounded electrodes in the one-dimensional and semi-classical WKB approximation in the

with respect to the tip and the surface densities of states. One can accomplish this by substituting $\xi = (E - eV/2)$ in equations 5 and 6. Then the differential conductivity is:

$$\begin{aligned} \frac{dI(S, V)}{dV} \cong \frac{Ae}{2} \cdot & \left[T'(S, \xi) \rho_s(\xi + eV/2) \rho_t(\xi - eV/2) \Big|_{\xi=eV/2} + \right. \\ & + T'(S, \xi) \rho_s(\xi + eV/2) \rho_t(\xi - eV/2) \Big|_{\xi=-eV/2} + \\ & + \int_{-eV/2}^{eV/2} T'(S, \xi) \frac{d\rho_s(\xi + eV/2)}{d\xi} \rho_t(\xi - eV/2) d\xi - \\ & \left. - \int_{-eV/2}^{eV/2} T'(S, \xi) \rho_s(\xi + eV/2) \frac{d\rho_t(\xi - eV/2)}{d\xi} d\xi \right], \end{aligned} \quad (8)$$

where

$$T'(S, \xi) \equiv T(S, V, E) = \exp \left[-2S \sqrt{\frac{2m}{\hbar^2} (\Phi - \xi)} \right] \quad (9)$$

is the appropriate presentation of the tunneling transmission probability (6).

Equation (8) is symmetric with respect to the surface and tip DOS. The $T'(S, eV/2)$ is a probability of tunneling to or from the tip Fermi level and the $T'(S, -eV/2)$ is a probability of tunneling to or from the sample Fermi level. As shown in Figure 1, at positive V (sample bias with respect to tip) $T'(S, eV/2) \gg T'(S, -eV/2)$. In this case the differential conductivity can be crudely estimated as:

$\rho_s(0) \cong \Delta\rho_s(0) \cong 0$, and the influence of the tip DOS is reduced at negative sample biases (11). Therefore, the sample density of occupied electronic states has a higher chance to reveal itself at negative tip-sample voltages despite the tunneling probability vanishing (cf. eqns. 8 and 11). This effect will be demonstrated by numerical simulations in Section IV.

III. Surface and Tip DOS Recovery by Normalization to Tunneling Probability.

The simplified equations (10, 11) show clearly the method of DOS deconvolution by using the differential conductivity. The best one can do is to normalize the dI/dV to $T'(S, eV/2)$ at positive sample bias and to $T'(S, -eV/2)$ at negative sample bias. Then the normalized differential conductivity will be proportional to the sample (10) or to the tip (11) DOS. As also seen from equations (10, 11), one may expect an influence of the first derivatives of the corresponding DOS on the recovered spectra. Nevertheless, all existing methods of DOS recovery from the tunneling spectroscopy data ultimately use this approach.^{17,18,21} The difference between various techniques is in the different methods of recovering the tunneling transmission probability.

III.1 Methods Based on Total Conductivity Utilization.

As was found empirically in many practical cases^{15,17,18,21} the total tunneling conductivity, I/V , follows reasonably well the desired normalization

probability or the corresponding differential conductivity, $\sigma_{const}(S, V)$. Hence, normalization of the differential conductivity to the total conductivity will not recover the constant input DOS, $\rho_s(\xi) = \rho_t(\xi) = 1$.

Significant deviation from the $\sigma_{const}(S, V)$ was also obtained for the modified total conductivity (Figure 2A). In this case double exponential weighting of the broadened and extended total conductivity was used to approximate the tunneling transmission probability (Refs. 17, 23):

$$\overline{I(S, V)/V} = \exp(a|V|) \int_{-\infty}^{\infty} \frac{I(S, \xi)}{\xi} \exp\left(-\frac{|eV - \xi|}{\Delta}\right) \times \exp(-a|\xi|) d\xi, \quad (14)$$

where a is the best parameter obtained during the $|I(S, V)|$ fitting by the exponential function $\exp(a|V|)$ and Δ is the broadening width. In all simulations $\Delta = 1$ eV was used.

Tunneling current versus voltage is always known in a limited voltage range. Hence, to avoid decay of the convoluted total conductivity in the proximity of the limits one should somehow extend the values of the tunneling current or total conductivity far beyond the limits of the voltage range. For variable tip-sample separation tunneling spectroscopy, Feenstra²³ has suggested extending the current values with constant value equal to the value of current at the largest measured voltage (the extension should be done separately for positive and negative voltages). For constant tip-sample separation spectroscopy it would be

where B_0 is the coefficient used at constant tip-sample separation.

DOS deconvolution accomplished by normalization of the differential conductivity to the modified total conductivity (Ref. 23) shows a systematic error in the case of variable tip-sample separation (Figure 2B). Therefore, in the framework of the one-dimensional WKB simulations, all techniques which employ the total conductivity for DOS recovery reveal significant deviation from the input tip and sample constant densities of states.

III.2 Method Based on Differential Conductivity Fitting.

The differential conductivity at a constant tip and surface DOS (eqn. 13) follows closely the appropriate tunneling probability at positive as well as at negative sample biases (Figure 1). Therefore, this function is a suitable one for DOS recovery by normalization (eqns. 10, 11).

If the sample Fermi level is located inside the band gap then $\rho_s(0) \ll \rho_t(0)$ and the tunneling differential conductivity should vanish at negative sample bias (eqn. 11). The symmetric tunneling probability function (13) is not appropriate for the fitting of this asymmetric differential conductivity. The situation can be fixed by employing an *asymmetric tunneling probability function*:

$$F(S, V) = A_T T'(S, eV/2) + A_S T'(S, -eV/2), \quad (17)$$

predicted that for these “up” and “down” atoms the difference in normal coordinate is about 0.5 Å. Therefore, for the STM probe placed above the dimer the density of states of the “up” atom of the asymmetric dimer are enhanced in the tunneling spectrum by a factor of 3 with respect to the density of states of the “down” one. In this analysis the decay factor has been assumed to be $k \approx 1 \text{ Å}^{-1}$ (Ref. 30).

Therefore, the fitting of experimental differential conductivity $\sigma(\varepsilon, V)$ by $\sigma_{\text{const}}(\varepsilon, V)$ and the following DOS recovery cannot be considered to be a quantitative procedure but rather should be viewed as a *qualitative* one. However, the proposed technique reveals the best DOS recovery at least in the frame of the WKB simulations.

IV. Numerical Simulation of the Electron Tunneling Spectroscopy. WKB approximation. One-Dimensional Case.

In the simulations presented in Figures 1-6, the following values were used: $B = 12 \text{ eV}^{-1/2}$ corresponding to the tip-surface separation of $\sim 11.7 \text{ Å}$, and the average tip-sample work function $\overline{\Phi} = 3 \text{ eV}$. Such values were obtained from the evaluation of tunneling spectra of Si(001)-(2x1) (Ref. 31), and are used here to better model the experimental situation.

In Figure 3 simulation for a sine tip and a sine surface DOS is presented. The periods of the tip and the sample DOS are 1 eV and 0.5 eV, respectively. As

To diminish the influence of tip DOS a new technique termed *Comparative Scanning Tunneling Spectroscopy (CSTS)* is proposed. The effectiveness of the approach is demonstrated in Figure 6. The deconvoluted DOS shown in Figure 5A was used as a reference spectrum. Two features were added to the input DOS of the *reference* sample on both sides of the band gap in order to get a *probe* sample input DOS (Figure 5B). The same tip DOS was used in both simulations. The difference of input sample densities of states (Fig. 6, dashed line) as well as tip DOS and output comparative spectra are presented in Figure 6.

The DOS averaged over macroscopic surface area can be obtained by traditional and inverse photoemission spectroscopies.³² Therefore, as soon as the reference DOS of the majority surface sites is known, the comparative analysis provides absolute spectra of surface electronic states with high spatial resolution. To get all advantages of the CSTS, both tunneling spectra (reference and probe) should be obtained with exactly the same tip. STM images are very sensitive to the tip condition and, hence, the later can be precisely monitored.

Employment of the tunneling probability function (13) for the comparative site specific tunneling spectroscopy on Si(001)-(2x1) revealed excellent reproducibility on a day-to-day and a tip-to-tip basis.³¹ Application of this comparative approach in the case of dI/dV normalization by the modified total conductivity is questionable, partially because of possible changes in local surface potential, tip-surface distance and effective tip area from one surface feature to

time, slow convergence and larger discrepancy caused by high correlation of the fitting parameters (eqns. 9, 17) was observed in some cases of highly modulated tip and sample densities of states. Such situations never occurred during experimental data evaluation.³¹

However, the data presented in Figure 7 can be considered as a *qualitative example* only. One should clearly realize that all simulations reported here were done in the one-dimensional and semi-classical WKB approximation. The proper test of the method has to be done in experiments or with a much more comprehensive theoretical approach (see refs. 15, 33).

V. Conclusion

A systematic study of surface DOS deconvolution from the tip-sample tunneling electron spectroscopy data was undertaken. The one-dimensional, semi-classical WKB approximation was used to simulate the electron tunneling between the metal tip and the surface. Using the simplest case of constant tip and sample densities of states (Fig. 2), it is shown that all current methods of DOS recovery based on the utilization of the total conductivity have a systematic error. In this paper a new technique for DOS deconvolution from tunneling spectroscopy data is introduced. *The differential conductivity normalized to its fit to the tunneling probability function* is employed as a method of recovering sample DOS. As shown by numerical simulations, this technique reveals a better deconvolution of

possible changes in local surface potential from one surface feature to another.

The influence of this parameter on the modified total conductivity is unknown.

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Figure 3. Tip influence on DOS deconvolution. Top graph: input sample (upper) and tip (lower) DOS. Bottom graph: solid line is the DOS deconvoluted by F normalization, dotted line is the DOS deconvoluted by normalization to the modified total conductivity at constant tip-sample separation $\overline{I/V}$, dash-dot line is the DOS deconvoluted by normalization to the modified total conductivity at variable tip-sample separation $\overline{I_m/V}$. Values used in the simulation are the same as for Figure 1.

Figure 4. Problem of sample DOS recovery below the Fermi level. Top graph: input sample (upper) and tip (lower) DOS. Bottom graph: solid line is the DOS deconvoluted by F normalization, dotted line is the DOS deconvoluted by normalization to the modified total conductivity at constant tip-sample separation $\overline{I/V}$, dash-dot line is the DOS deconvoluted by normalization to the modified total conductivity at variable tip-sample separation $\overline{I_m/V}$. Values used in the simulation are the same as for Figure 1.

Figure 5A. Deconvolution of the simple semiconductor sample DOS. Top graph: input sample (upper) and tip (lower) DOS. Bottom graph: solid line is the DOS deconvoluted by F normalization, dotted line is the DOS deconvoluted by normalization to the modified total conductivity at constant tip-sample separation

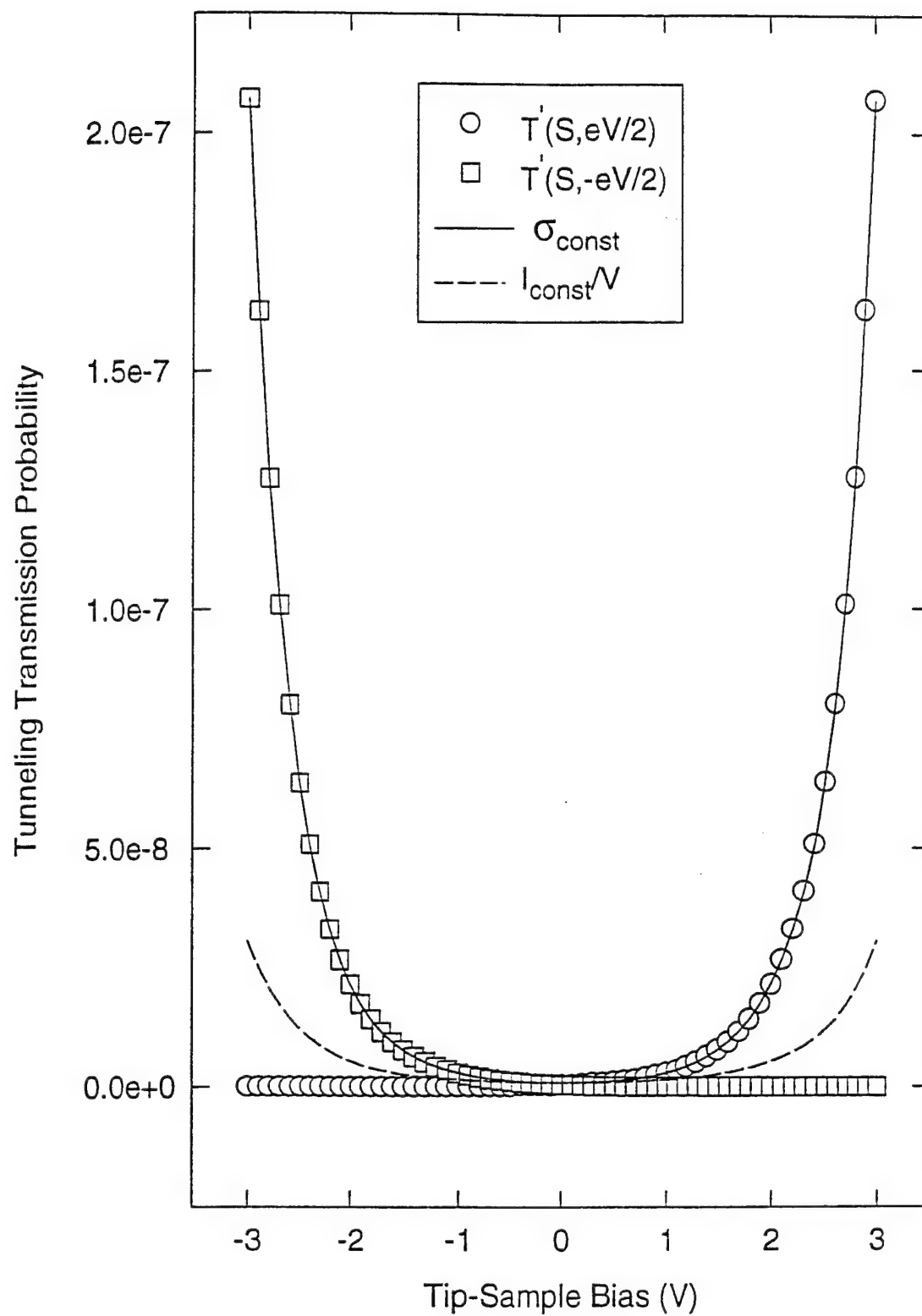
variable tip-sample separation $\overline{I_m/V}$. Values used in the simulation are the same as for Figure 1.

Figure 7A. Work function (WF) and tip-sample distance (TSD) recovery in the case of the simple semiconductor sample DOS. The tunneling current vs. tip-sample bias dependencies were fitted by the tunneling probability function (17). Top graph: solid line is the dependence of recovered tip-sample distance on input work function, dotted line is the input tip-sample distance. Bottom graph: solid line is the dependence of the recovered work function on input work function, dotted line is the input work function.

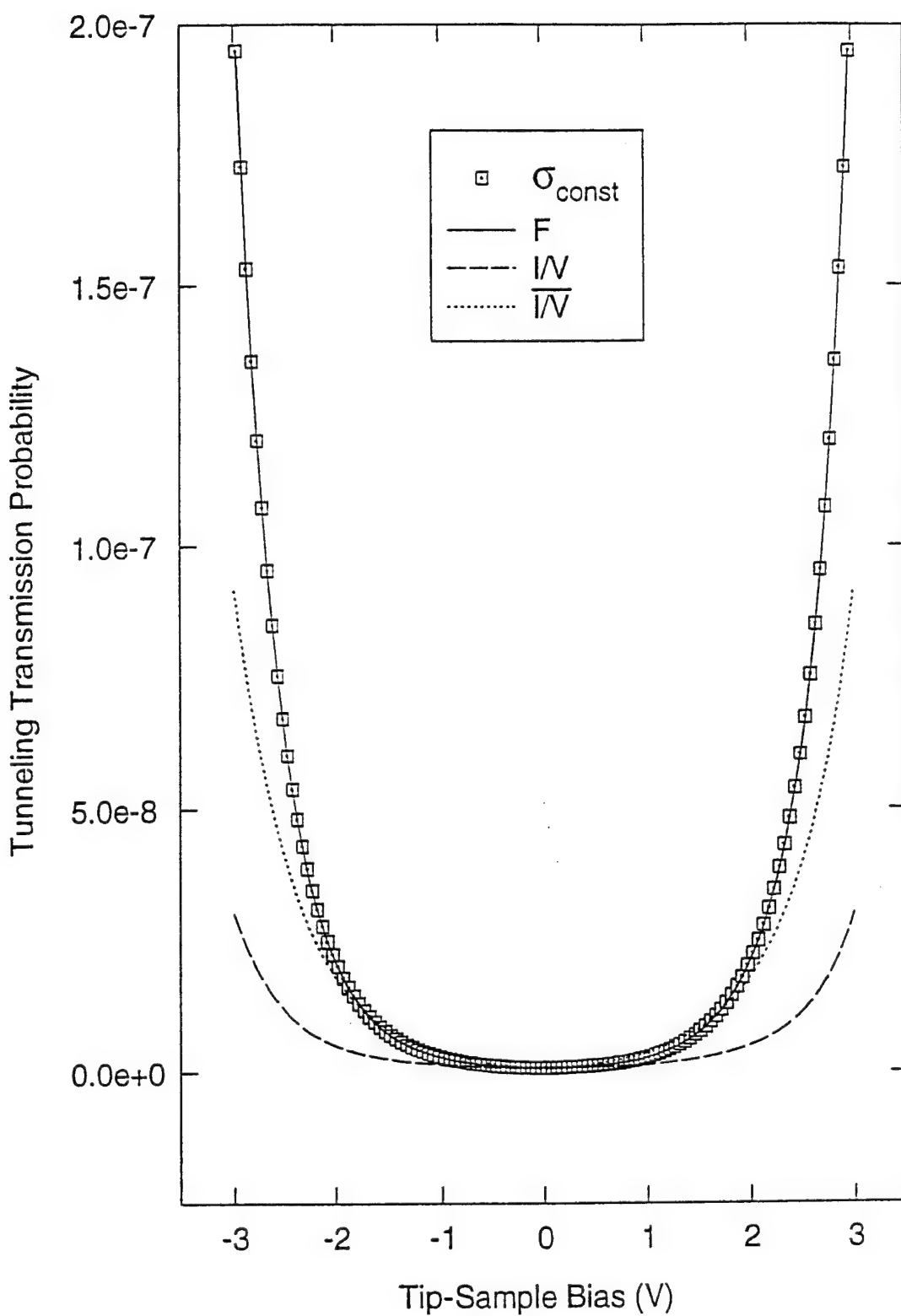
Figure 7B. Work function (WF) and tip-sample distance (TSD) recovery in the case of the simple semiconductor sample DOS. The tunneling current vs. tip-sample bias dependencies were fitted by the tunneling probability function (17). Top graph: solid line is the dependence of the recovered work function on input tip-sample distance, dotted line is the input work function. Bottom graph: solid line is the dependence of the recovered work function on input work function, dotted line is the input work function.

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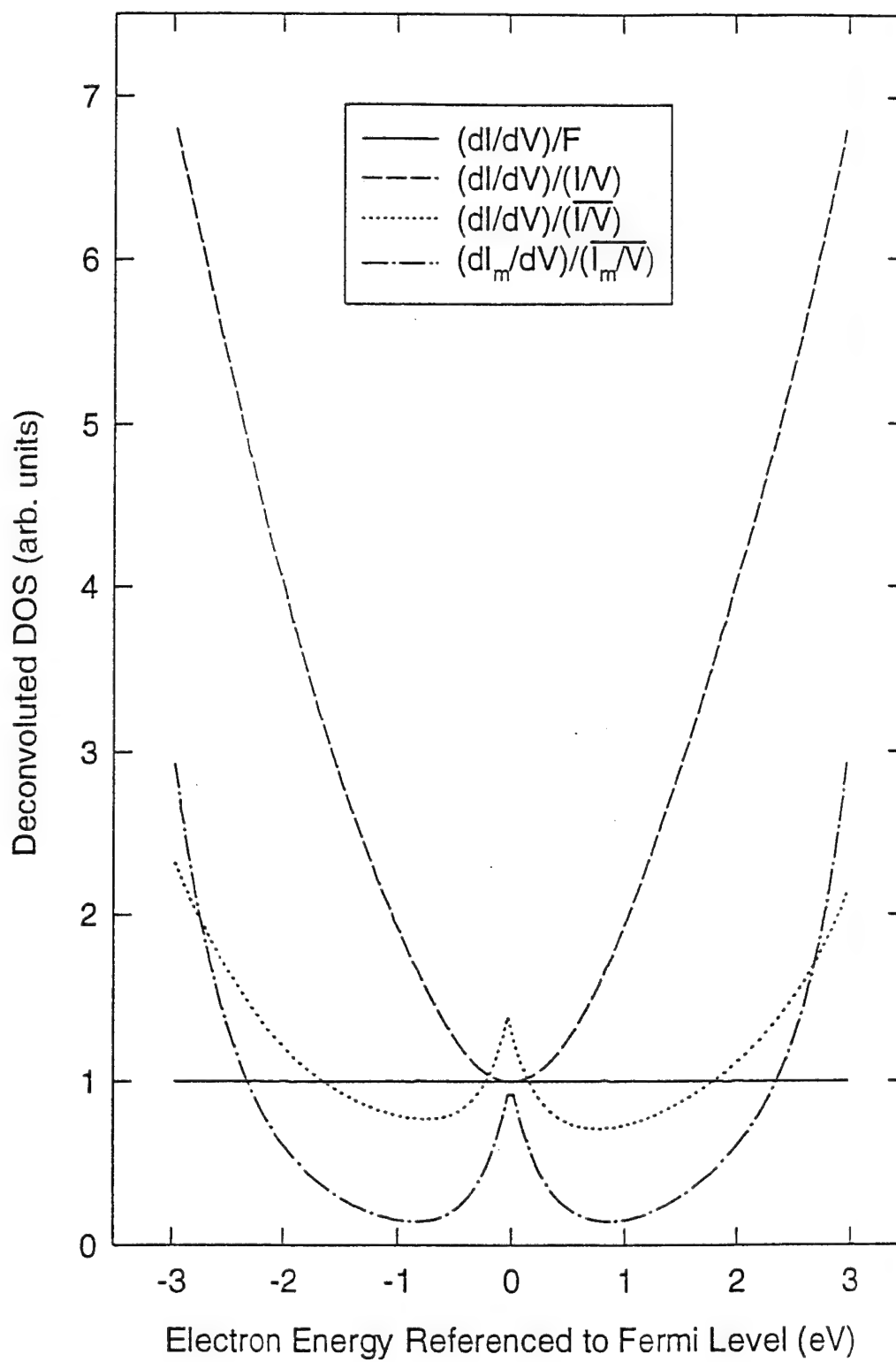
Tunneling Transmission Probability.



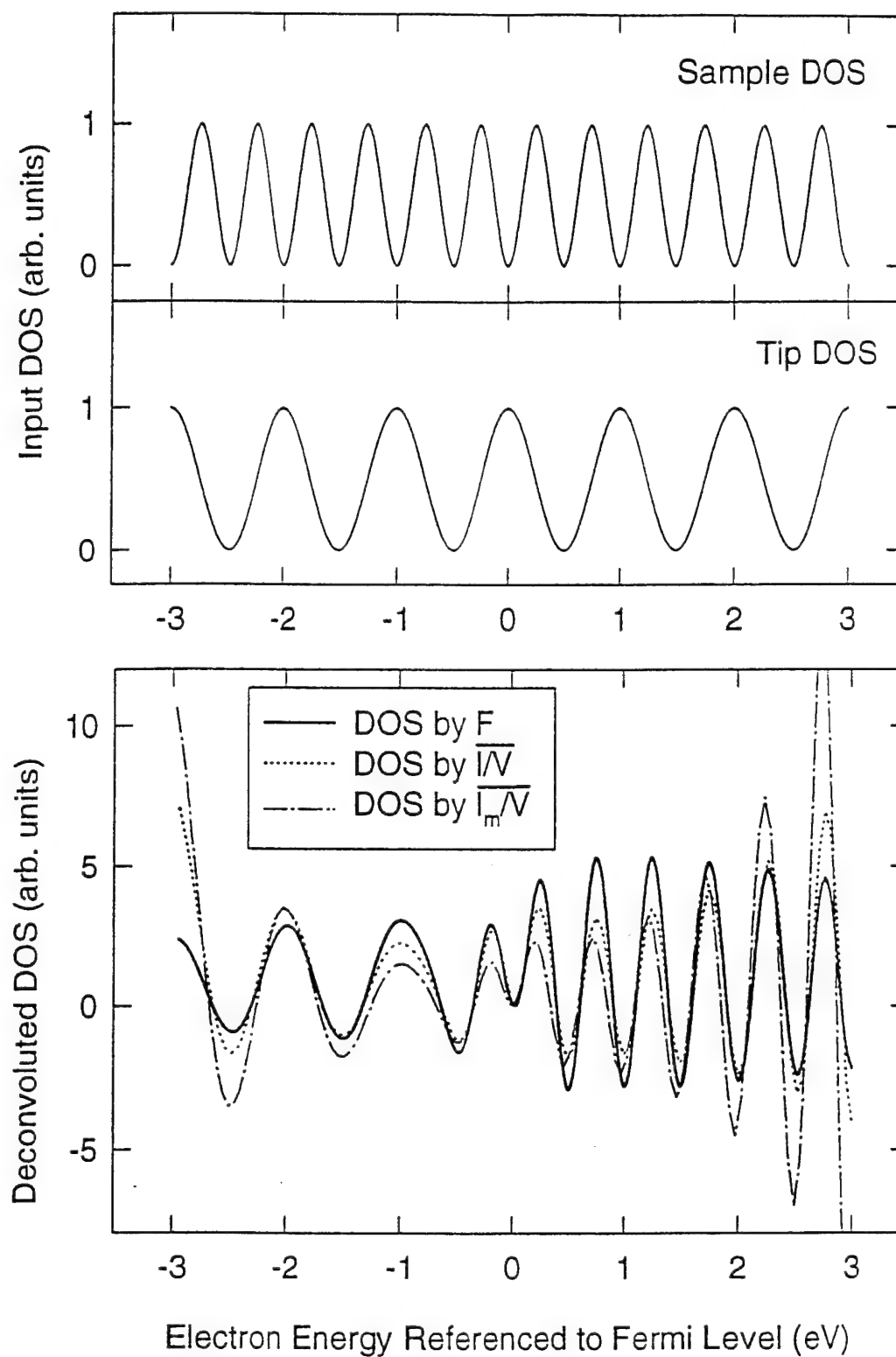
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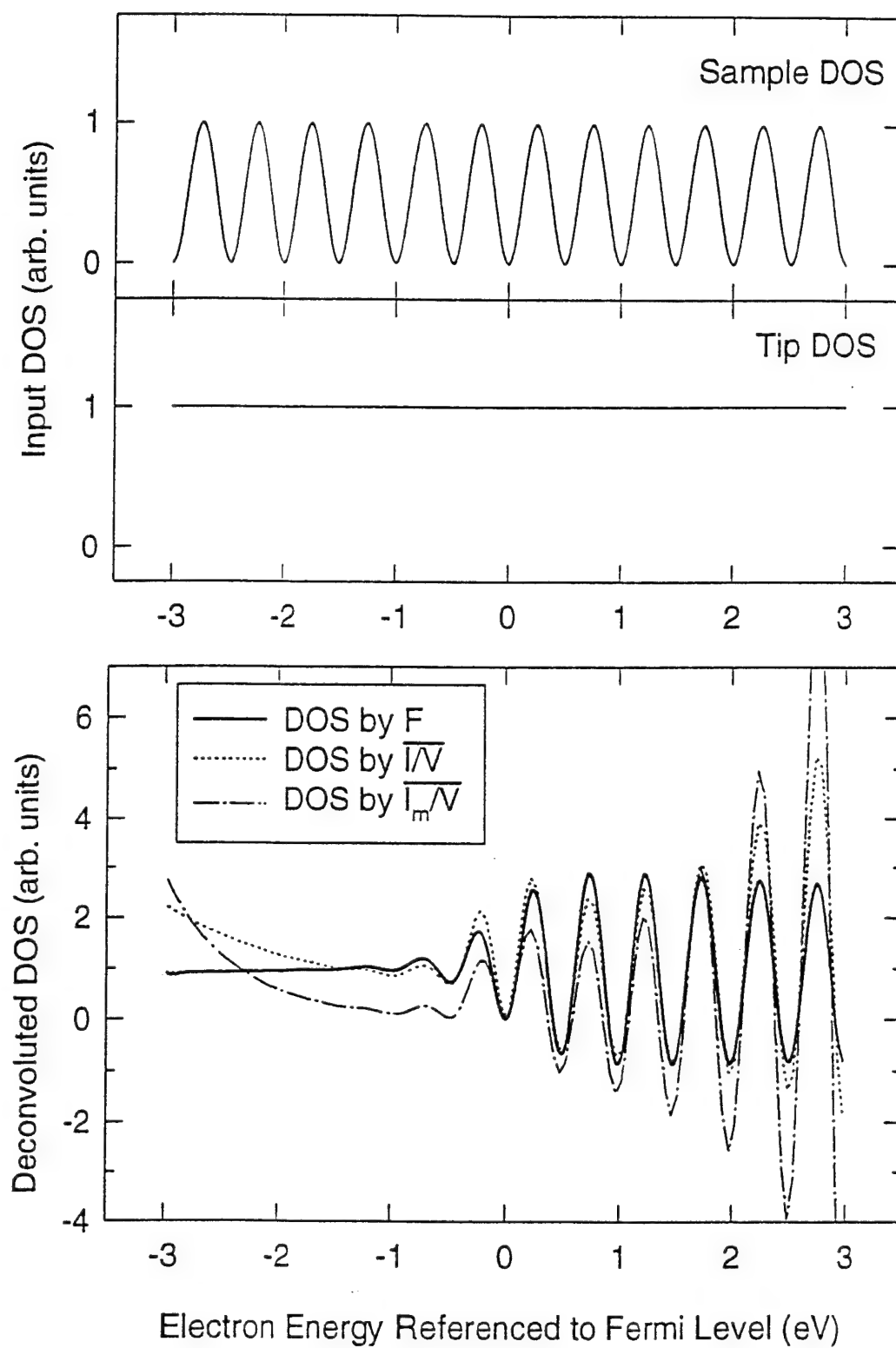
Deconvoluted DOS.



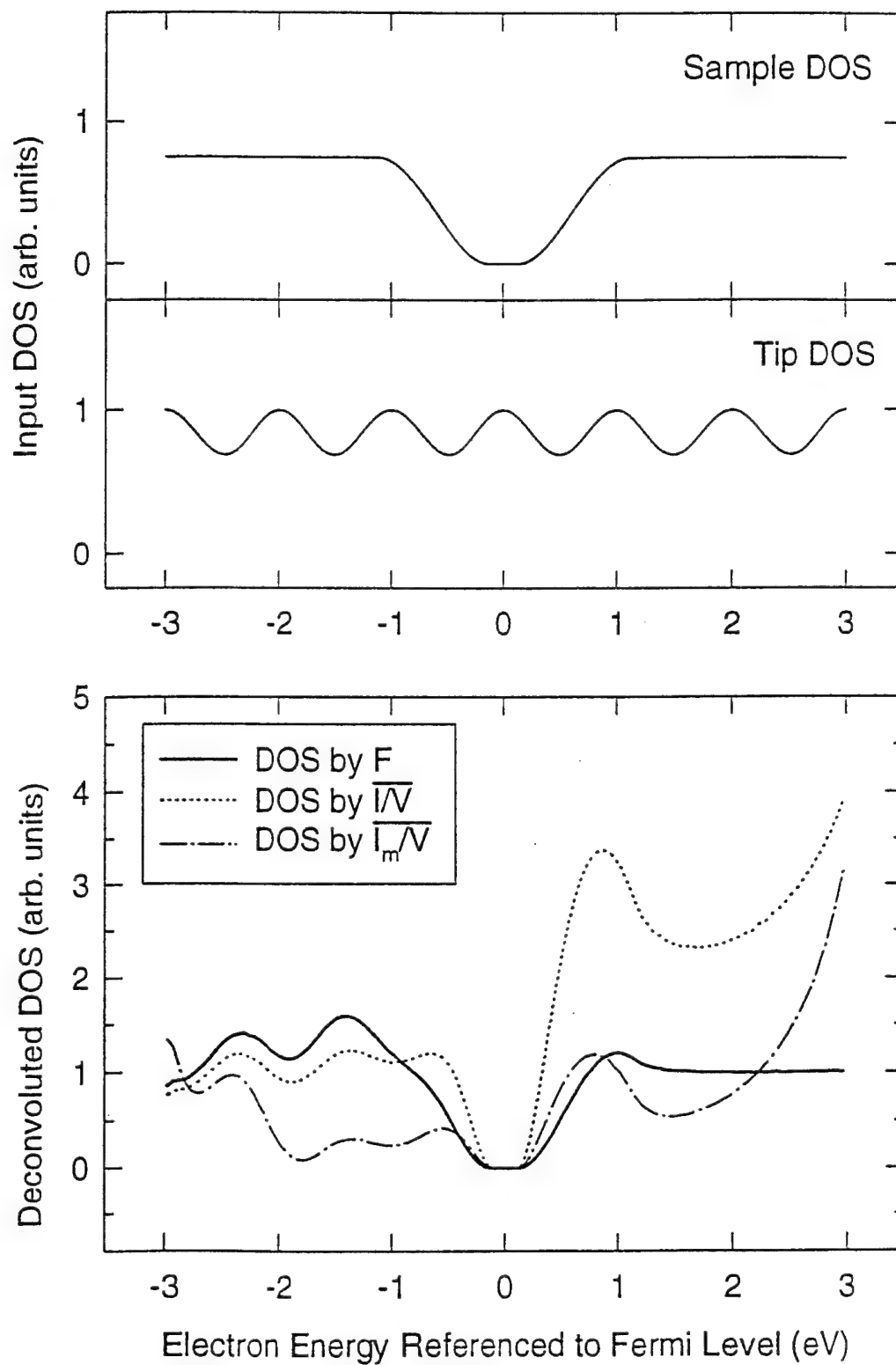
Tip Influence on DOS Deconvolution.



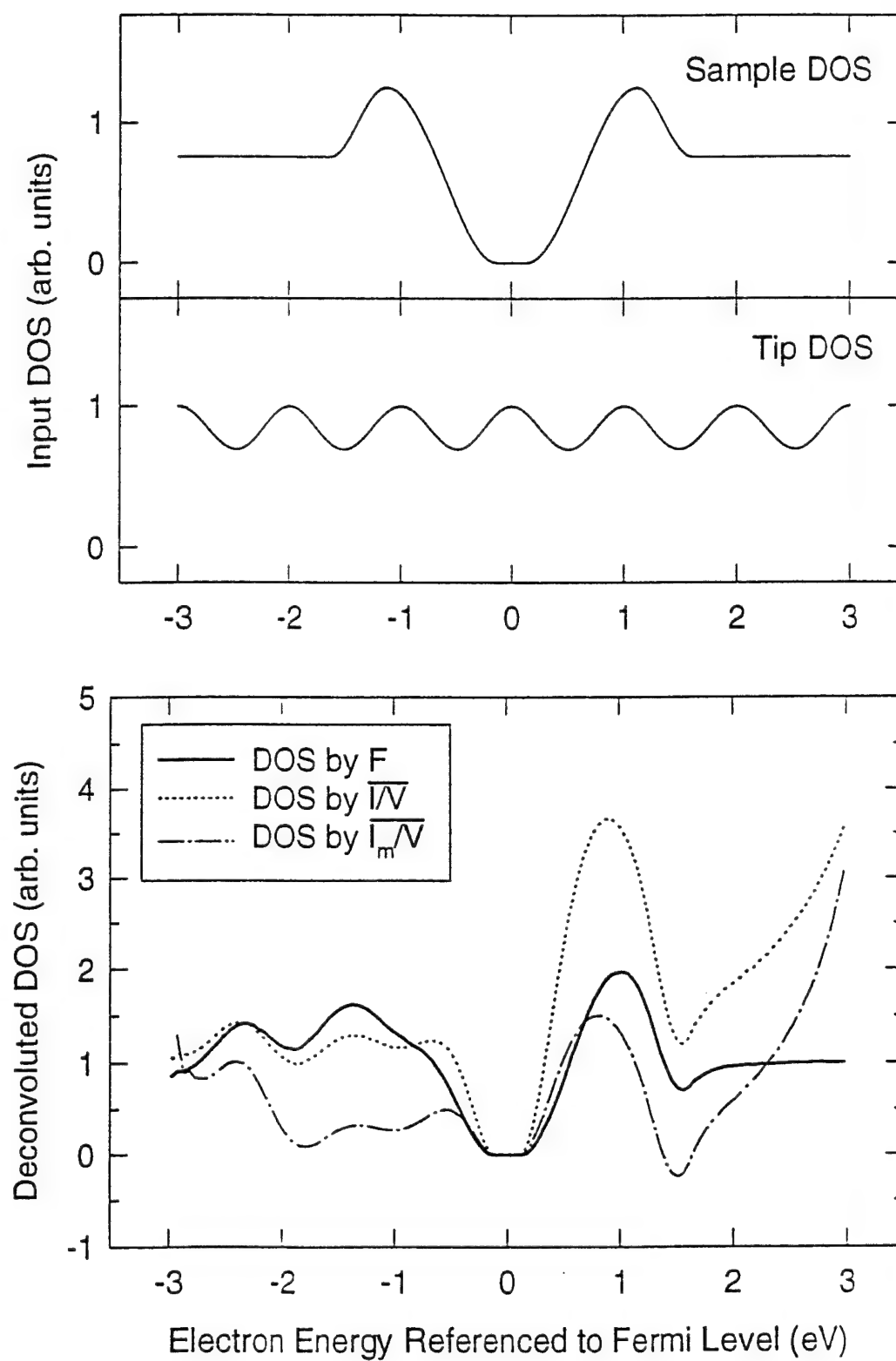
Problem of Sample DOS Recovery Below Fermi Level.



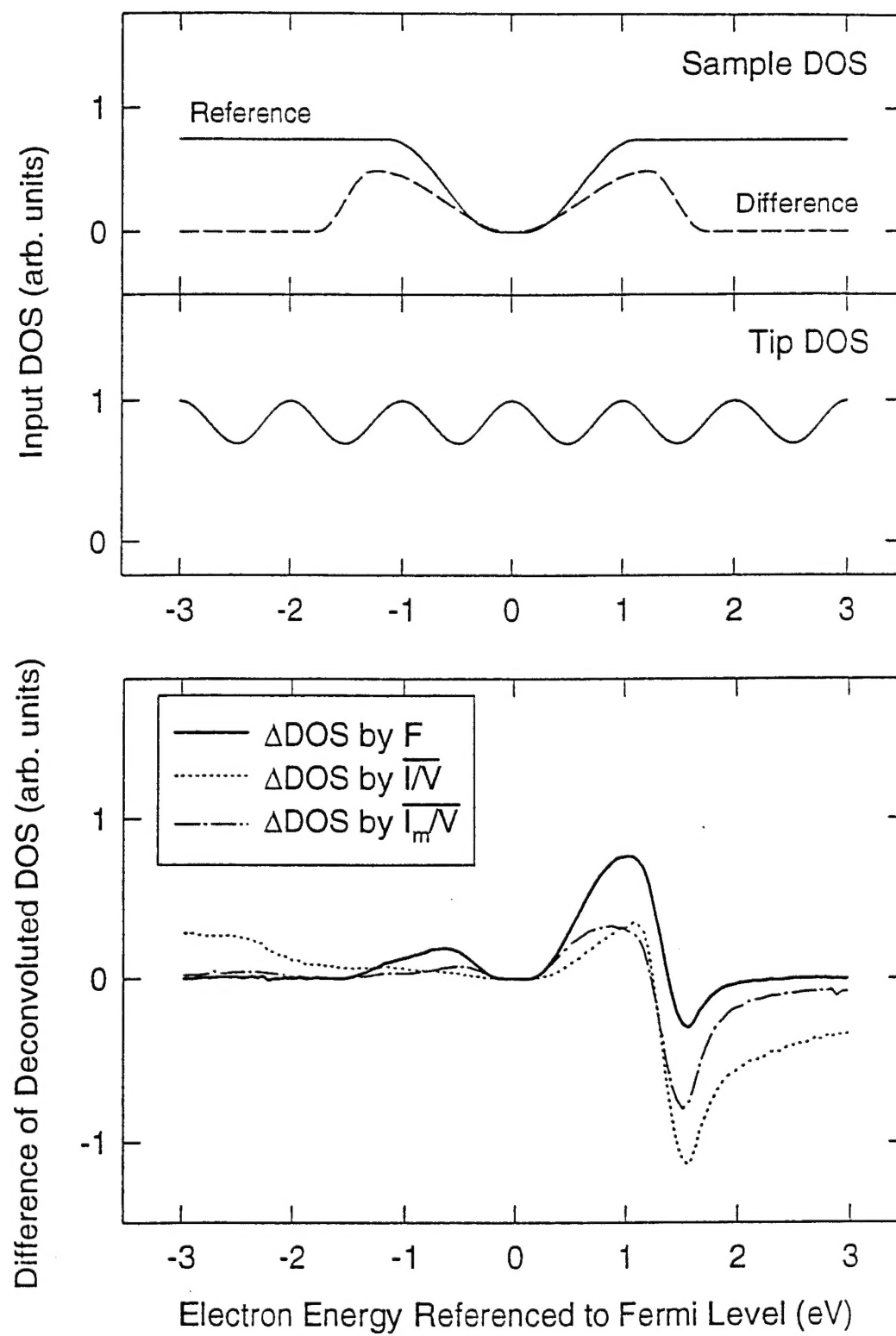
Deconvolution of Simple Semiconductor DOS.



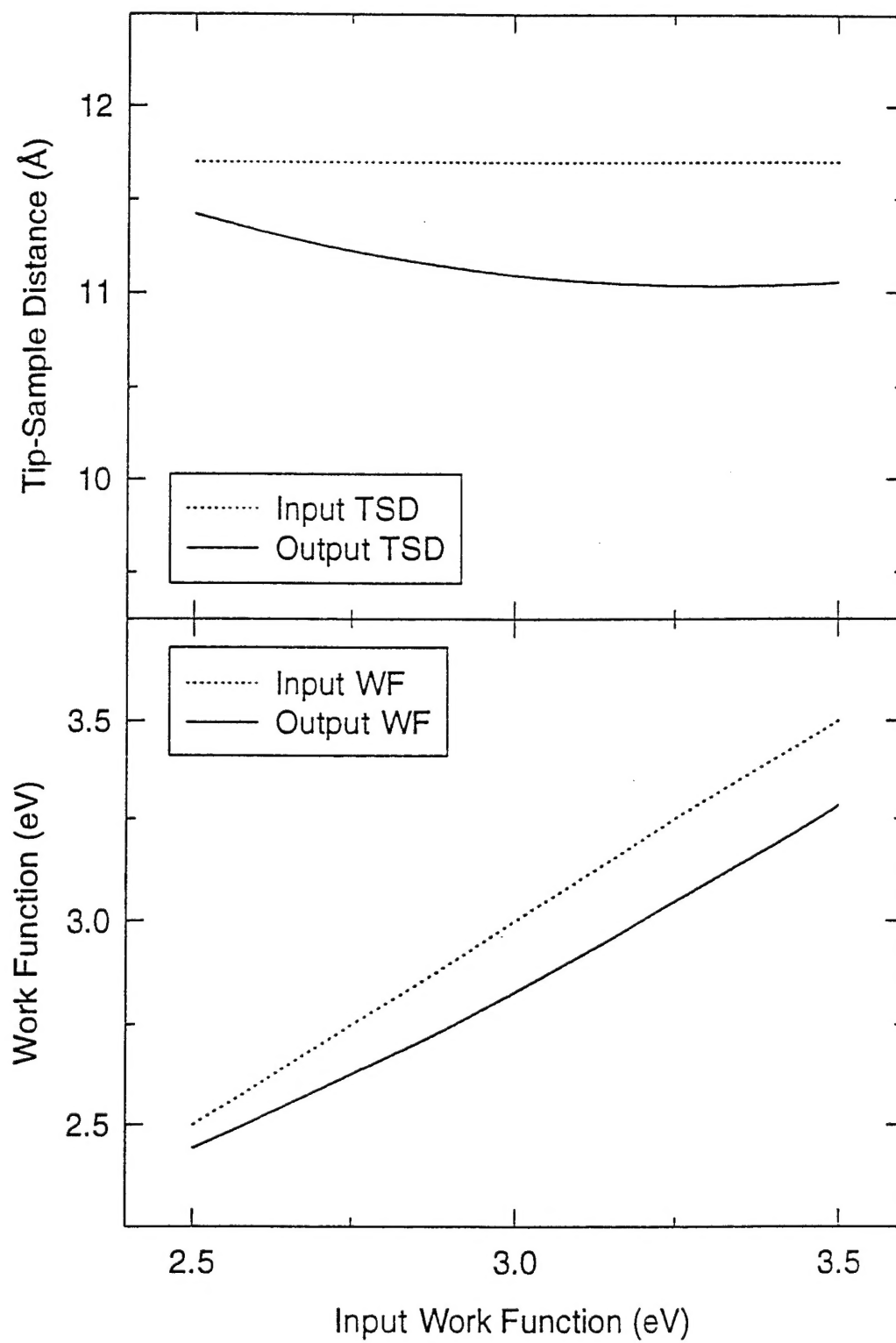
Deconvolution of Augmented Semiconductor DOS.



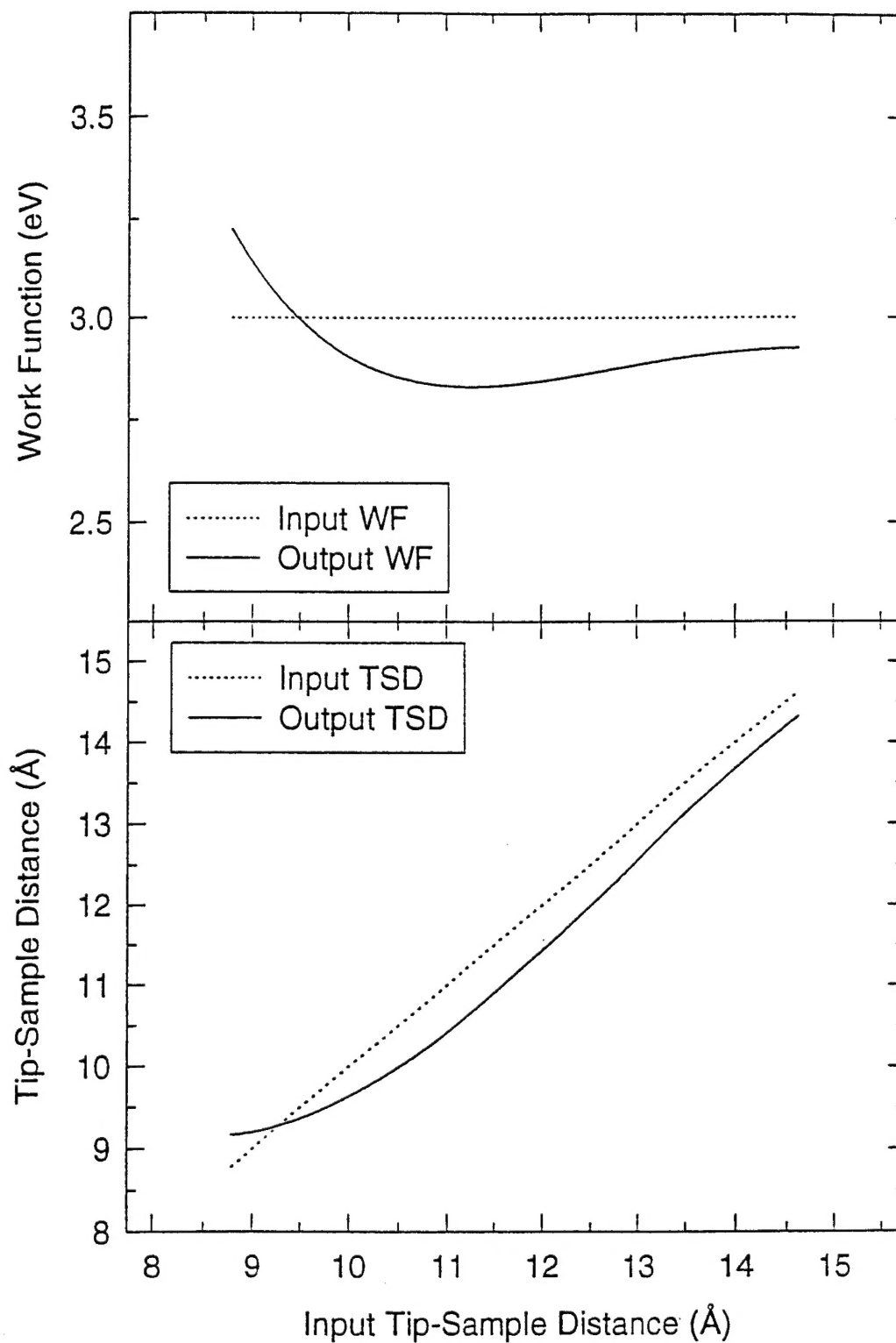
Example of Comparative Tunneling Spectroscopy.



Work Function and Tip-Sample Distance Recovery. Simple Semiconductor Sample DOS.



Tip-Sample Distance and Work Function Recovery. Simple Semiconductor Sample DOS.



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